

# A liquid-liquid extraction system suitable for studying the chemistry of rutherfordium with SISAK

D. Poláková<sup>1</sup>, G. Skarnemark<sup>2</sup>, J. Alstad<sup>1</sup>, L. Stavsetra<sup>1</sup>, L. Zheng<sup>1</sup>, J.P. Omtvedt<sup>1</sup>

<sup>1</sup> University of Oslo, Norway

<sup>2</sup> Chalmers University of Technology, Sweden

darina.polakova@kjemi.uio.no

## Introduction

A general presentation of the on-line liquid-liquid extraction system SISAK is given in another presentation to the TAN'03 conference (J.P. Omtvedt and coworkers). The presentation given here describes a system suitable for the investigation of the chemistry of rutherfordium. This work is in progress, and *the results presented are preliminary*.

In 2000 and 2001 a pilot SISAK chemistry-experiment with the transactinide rutherfordium was performed at the Lawrence Berkeley National Laboratory (LBNL) in Berkeley, USA. In this experiment it was proved that it was possible to extract and detect the ~4 s transactinide <sup>257</sup>Rf with SISAK [1].

The <sup>257</sup>Rf activity was produced with a 235 MeV <sup>50</sup>Ti<sup>12+</sup> beam from the LBNL 88-inch cyclotron hitting a rotating <sup>208</sup>Pb target. The recoils from the target were separated in the Berkeley Gas-filled Separator (BGS). After this, the recoils were transferred from the BGS focal plane into a gas jet by using a then newly constructed Recoil Transfer Chamber (RTC). The activity is transported to the SISAK system with a gas jet. First, in the degasser step, the activity is transferred from gas phase to an aqueous phase, then the activity is extracted with a suitable extraction agent into the organic phase. By carefully selecting the conditions and extraction agents, properties like valence state, charge state, the number and type of ligands can be investigated. The extracted rutherfordium (in the organic phase) is then mixed with a scintillator cocktail and pumped through Liquid Scintillation (LS) detectors. The amount of rutherfordium remaining in the aqueous state can currently not be measured, but must be calculated from measurements of how much activity is entering the SISAK system. An overview of the complete setup is shown in Fig. 1.

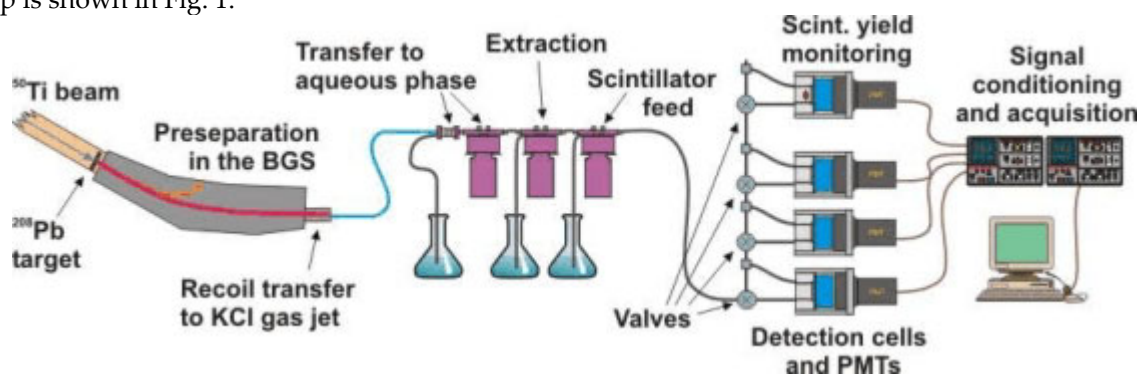


Figure 1. Overview of the complete SISAK setup for transactinide experiments.

As reported in one of our other TAN'03 contributions (J.P. Omtvedt and coworkers), the chemistry system (extraction with dibutyl-phosphate from nitric acid) used in the 2000 and 2001 experiments was not found suitable for detailed chemistry studies of Rf. Therefore the still on-going work reported here was initiated to find a suitable system for investigation of the liquid-phase chemistry of Rf with SISAK.

## Search for a suitable extraction systems for Rf experiments

A chemical system, i.e. the concentration and flow rates of the reagents, and the solvents used, must meet a number of criteria to be suitable for a SISAK type of chemical investigation of transactinides:

1. Good separation of Zr and Hf. This is because it is of interest to be able to classify the behaviour of rutherfordium as either Zr or Hf like.
2. No adverse effects (like adsorption) between the chemicals used and the construction material of the SISAK equipment.
3. The complexation and extraction rates must be fast. The contact time between the two liquid phases is between 0.5 and 0.1 s. Therefore the reaction rate must be fast enough to at least approach equilibrium.
4. No (or only small) interference with the liquid scintillation used to detect the  $\alpha$  activity can be allowed. Otherwise the detection will be very uncertain or even impossible.

In works by Yagodin and coworkers [2-4], a liquid-liquid extraction system based on extraction of carrier amounts from dilute  $\text{H}_2\text{SO}_4$  with trioctylamine (TOA) was reported. A good separation between Zr and Hf is achieved. Thus, this seemed like a good starting point for an extraction system to investigate the chemical properties of rutherfordium. A schematic diagram of how this extraction system is setup with SISAK is shown in Fig. 2. Results from our initial tests with carrier free amounts with such a system are shown in Fig. 3.

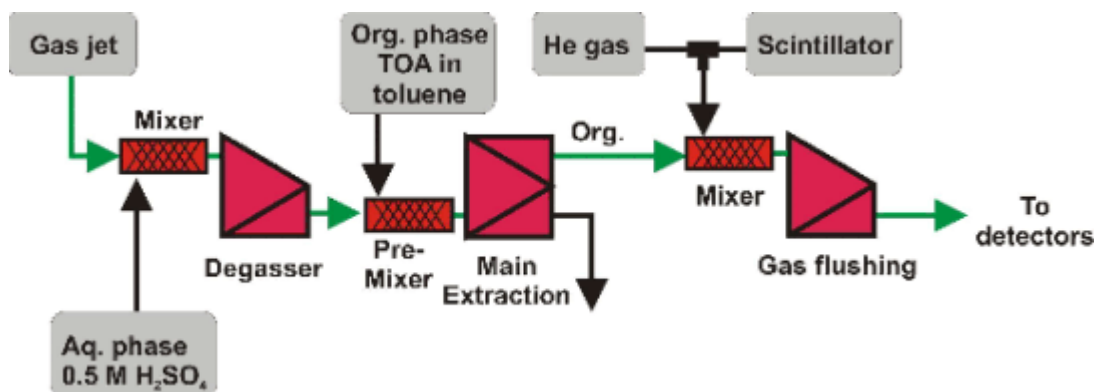


Figure 2. SISAK setup for extraction of carrier free amounts of Zr, Hf and Rf with TOA from sulphuric acid.

The data for Fig. 3 was obtained from on-line experiments at the Oslo Cyclotron Laboratory (OCL). A  $\sim 200$  nA 45 MeV  $^3\text{He}^{2+}$  beam was used on a combined Sr/Yb target (no isotopic enrichment for either element). The reaction products recoil out of the target and are caught on KCl-aerosol particles in a helium gas-jet. The gas-jet transports the activity in a few seconds to a chemistry lab through a 20 m capillary. Thus, we obtain 14 s  $^{87\text{m}}\text{Zr}$  and 3.25 m  $^{169}\text{Hf}$  activity for on-line SISAK experiments. The advantage of this approach, compared to using off-line produced activity dissolved directly in one of the liquid phases, is that the conditions are nearly identical to those encountered in rutherfordium experiments.

As can be seen from the right-hand panel in Fig. 3, the separation factor between the distribution ratio for Zr and Hf is on average 5.7. This is high enough to satisfy condition no. 1 in the list above. Experiments to optimize this separation with respect to reagent concentrations are under way and the results will be reported at the TAN'03 conference.

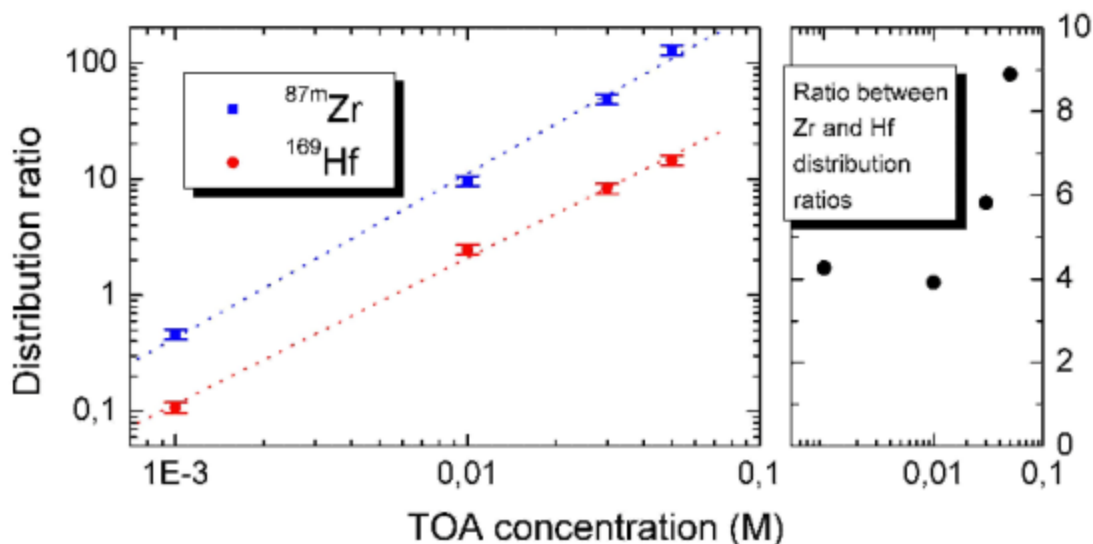


Figure 3. On-line extraction curves for Zr and Hf using 0.5 M  $\text{H}_2\text{SO}_4$  aqueous phase and various TOA concentrations in toluene as organic phase.

In extended tests of this chemistry we found no adsorption or other unwanted effects between the chemicals used and the SISAK equipment. Thereby, condition no. 2 above is met. By comparing batch extractions performed at equilibrium conditions and on-line extraction, the difference was found to be small. This indicates that condition no. 3 is also met, a slow system would not have approached the equilibrium distribution-ratio. Finally, the quenching after contacting the organic phase with the sulphuric-acid aqueous phase was found small enough to not be of any major concern. Thus, the extraction system described here looks very promising as a system for future chemical studies of rutherfordium. More details and results will be presented at the TAN'03 conference.

## References

- [1] J.P. Omtvedt *et al.* J. Nucl. Radiochem. Sci. **3**, 121 (2002).
- [2] G.A. Yagodin, A.M. Chekmarev, and L.M. Vladimirova. Russ. J. Inorg. Chem. **14**, 838 (1969).
- [3] G.A. Yagodin, O.A. Sinegribova, and A.M. Chekmarev. In: Proc. Int. Solvent Extraction Conf., 1971, The Hague.
- [4] G.A. Yagodin *et al.* Russ. J. Inorg. Chem. **27**, 448 (1982).